

Quantum Stochastic Synchronization

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We study within the spin-boson dynamics the synchronization of quantum tunneling with an external periodic driving signal. As a main result we find that at a sufficiently large system-bath coupling strength (Kondo parameter $\alpha > 1$) the thermal noise plays a constructive role in yielding both a frequency and a phase synchronization in a symmetric two-level system. Such riveting synchronization occurs when the driving frequency supersedes the zero temperature tunneling rate. As an application evidencing the effect, we consider a charge transfer dynamics in molecular complexes.

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The phenomenon of synchronization of nonlinear classical systems with external driving signals, possibly even in the presence of randomness, has increasingly been gaining importance and growing interest over the last decade [1, 2, 3, 4]. A particular intriguing example is crowd synchrony when pedestrians fall into steps with the intrinsic vibrations of a footbridge [5]. Physically related to the synchronization phenomenon is the phenomenon of Stochastic Resonance [6] where the presence of noise can manifestly boost the transduction of an information-carrying, weak signal. This effect is enduring ongoing vitality in diverse fields that span from physics to the life sciences. This fascinating phenomenon has notably been generalized into the quantum regime only recently [7, 8, 9], where its experimental realization on the level of a nanomechanical quantum memory element is very feasible [10]. The extension of noise-induced synchronization into the realm of quantum physics, however, has not been considered thus far. This latter task presents a challenge which, apart from basic academic interest, also comprises great potential for nanoscience with promising applications ranging from quantum control to quantum information processing.

With this work we undertake a first step in this direction. We consider the paradigm of a driven, symmetric quantum two-level system that is coupled to an Ohmic thermal bath with an exponential cutoff [9, 11]. When operating within the overdamped regime, we successfully reveal the existence of a noise-assisted *quantum synchronization*. There are two relevant energetic parameters characterizing the system-bath coupling: the reorganization energy λ and the cutoff energy $\hbar\omega_c$ which is associated to the spectral width of the medium modes which couple to the tunneling particle. The two parameters can be combined into the dimensionless coupling strength $\alpha = \lambda/(2\hbar\omega_c)$, i.e., the Kondo parameter. A third relevant energy scale constitutes the tunneling energy $\hbar\Delta$. The most relevant regime for our purposes corresponds to $\hbar\Delta \ll \hbar\omega_c, \lambda$ and $\alpha > 1$.

Dissipative quantum tunneling changes radically the physics of synchronization. At zero temperature, the system can only tunnel towards its lowest energy state when

a biasing dc-signal is applied. As the bias periodically changes its sign due to the action of a driving field, tunneling makes the particle move periodically towards its corresponding lowest energy state, as long as the driving period is much longer than the typical tunneling time. Consequently, one expects that the system synchronizes with a driving periodic rectangular signal. By contrast, in the absence of classical thermal noise, synchronization in a overdamped bistable system driven by rectangular subthreshold signals does not occur, as the overbarrier transitions do not exist.

Two interesting questions now emerge: What is the effect of quantum noise at finite temperature for synchronization? How does the time scale of the external driving period impact this synchronization behavior? Quantum bath fluctuations at finite temperature surely will disturb the above mentioned perfect synchronization of the tunneling events as imposed by the rocking driving field. At the same time, when the driving period becomes shorter than the tunneling time, the zero temperature synchronization will also be weakened. Nonetheless, it might very well be possible that finite temperature quantum noise promotes and assists synchronization when the driving period decreases. We demonstrate below that this indeed is the case.

To start, we consider the following archetype model of a dissipative, driven two-level system (TLS) [9, 11, 12, 13]:

$$\begin{aligned} \hat{H}(t) = & \frac{1}{2}\epsilon(t)\hat{\sigma}_z + \frac{1}{2}\hbar\Delta\hat{\sigma}_x + \frac{1}{2}\hat{\sigma}_z \sum_j \kappa_j(b_j^\dagger + b_j) \\ & + \sum_j \hbar\omega_j(b_j^\dagger b_j + \frac{1}{2}). \end{aligned} \quad (1)$$

Herein, the operators $\hat{\sigma}_z$ and $\hat{\sigma}_x$ denote the standard Pauli operators, $\epsilon(t)$ is a time-dependent energy bias, and $\hbar\Delta$ in (1) is the tunneling matrix element. The boson operators b_j^\dagger and b_j correspond to normal mode oscillators of the thermal bath with frequencies ω_j . The stochastic influence of the quantum thermal bath is captured by an *operator* random force $\hat{\xi}(t) = \sum_j \kappa_j(b_j^\dagger e^{i\omega_j t} + b_j e^{-i\omega_j t})$. It can be characterized by the spectral den-

sity $J(\omega) = (\pi/\hbar) \sum_j \kappa_j^2 \delta(\omega - \omega_j)$. We assume that $J(\omega)$ acquires the Ohmic form with an exponential cutoff, $J(\omega) = 2\pi\hbar\alpha\omega e^{-\omega/\omega_c}$. Due to the Gaussian statistics of a harmonic bath, the statistical properties of the quantum noise become completely determined by its equilibrium autocorrelation function $\langle \hat{\xi}(t)\hat{\xi}(0) \rangle_T = (\hbar/\pi) \int_0^\infty J(\omega) [\coth(\hbar\omega/2k_B T) \cos(\omega t) - i \sin \omega t] d\omega$. The driven spin-boson Hamiltonian (1) describes many situations, such as, e.g., the electron transfer (ET) in a molecular dimer in azurin crystals [14]. Then the low-frequency molecular vibrations provide the bath and the time-dependent energy bias is given by $\epsilon(t) = re\mathcal{E}(t)$, where r is the tunneling distance, e denotes the charge transferred, and $\mathcal{E}(t)$ is the time-dependent, applied electric field.

In the incoherent tunneling regime, the populations of the localized states obey a nonstationary, Markovian dynamics. In the presence of a time-dependent bias, this description holds true for Ohmic friction at an arbitrary temperature if the tunneling coupling remains small, i.e. $\Delta \ll \omega_c$, and the coupling to the heat bath is sufficiently strong, $\alpha > 1/2$ [13]. The populations $p_\pm(t) = (1 \pm \langle \sigma_z(t) \rangle_T)/2$ then obey the balance equations [9, 11, 15]

$$\dot{p}_\beta(t) = W_{-\beta}(t)p_{-\beta}(t) - W_\beta(t)p_\beta(t) \quad (2)$$

with $\beta = \pm$, where the time-dependent relaxation rates $W_\beta(t)$ are given by

$$W_\pm(t) = \frac{1}{2}\Delta^2 \int_0^\infty d\tau \exp[-Q'(\tau)] \times \cos \left[Q''(\tau) \mp \frac{1}{\hbar} \int_{t-\tau}^t \epsilon(t') dt' \right] \quad (3)$$

within the Golden Rule approximation. The functions $Q'(t)$ and $Q''(t)$ in (3) denote the real and imaginary parts of

$$Q(t) = \frac{i\lambda t}{\hbar} + \frac{1}{\hbar^2} \int_0^t dt_1 \int_0^{t_1} dt_2 \langle \hat{\xi}(t_2)\hat{\xi}(0) \rangle_T, \quad (4)$$

wherein $\lambda = \int_0^\infty d\omega J(\omega)/(\pi\omega)$ is the bath reorganization energy [15]. Here, $\lambda = 2\alpha\hbar\omega_c$ and the function $Q(t)$ can be evaluated in closed analytical form [13, 16], reading

$$Q'(t) = 2\alpha \ln \left\{ \sqrt{1 + \omega_c^2 t^2} \frac{\Gamma^2(1 + \kappa)}{|\Gamma(1 + \kappa + i\omega_T t)|^2} \right\} \quad (5)$$

$$Q''(t) = 2\alpha \arctan(\omega_c t). \quad (6)$$

In Eq. (5), $\Gamma(z)$ denotes the Gamma function, $\omega_T = k_B T/\hbar$, and $\kappa = \omega_T/\omega_c$. Note that in the limit of an *adiabatic* driving varying on a time-scale \mathcal{T} such that $\omega_c \mathcal{T}, \alpha \omega_T \mathcal{T} \gg 1$, the time-dependent transition rates $W_\pm(t)$ follow the *instantaneous* value of the bias $\epsilon(t)$. In this limit, which is assumed throughout in the following, the relaxation rates $W_\pm(t)$ obey the Boltzmann relation $W_+(t) = e^{\epsilon(t)/k_B T} W_-(t)$. Furthermore, in the high-temperature limit $k_B T \gg \hbar\omega_c$, Eq. (3) reduces to a generalized Marcus-Levich-Dogonadze form, i.e., $W_\pm(t) =$

$(\pi/2)\hbar\Delta^2/\sqrt{4\pi\lambda k_B T} \exp[-(\pm\epsilon(t) - \lambda)^2/4\lambda k_B T]$. For $k_B T \leq \hbar\omega_c$, explicit analytical expressions for the rates are generally not available, except for $T = 0$ [13]; typically, those must be determined numerically from Eq. (3).

Using the Marcus-Levich-Dogonadze formula for an undriven molecular system, one can estimate the relevant parameter values. In particular, for ET occurring in azurin dimer the values are $\lambda = 0.25$ eV and $\hbar\Delta = 5 \cdot 10^{-6}$ eV [14]. In molecular systems, the cutoff frequency of low-frequency molecular vibrations ranges between $\hbar\omega_c = 5 - 20$ meV. Choosing $\hbar\omega_c = 12.5$ meV yields $\alpha = 10$ corresponding to a strongly incoherent, overdamped tunneling dynamics.

It is worth noting that the present incoherent limit for the tunneling dynamics of a driven, dissipative TLS allows for an effective *quasiclassical* interpretation in terms of a classical random telegraph process which is inhomogeneous in time. Its transition rates, however, are governed by the manifestly *quantum expressions* detailed in Eq. (3). As such, our setup mimics the quantum analogue of a noisy, classical synchronization behavior elaborated in Refs. [17, 18, 19]. A thought-experimental setup is that of a particle tunneling between two localized states at random times subject to an external periodic rectangular field with amplitude \mathcal{E}_0 and period \mathcal{T} (frequency $\Omega = 2\pi/\mathcal{T}$). One counts the number of jumps $N(t, t_0)$ within a time window $[t_0, t]$. Following Refs. [17, 18, 19], we introduce the random phase $\phi(t, t_0) = \pi N(t, t_0)$, which increases by π at each switching event (two subsequent switches correspond to a 2π -cycle with random duration). We define the mean frequency and diffusion coefficients associated to the phase process as $\overline{\Omega}_{\text{ph}} := \lim_{t \rightarrow \infty} \langle \phi(t, t_0) \rangle / (t - t_0)$ and $2\overline{D}_{\text{ph}} := \lim_{t \rightarrow \infty} [\langle \phi^2(t, t_0) \rangle - \langle \phi(t, t_0) \rangle^2] / (t - t_0)$, respectively. To evaluate these quantities, one can consider the joint probability $P_{\beta,n}(t)$ to be in the state β at time t and to have n jumps within the time interval $[t_0, t]$. These probabilities can be obtained by integrating the multi-time probability densities of the corresponding stochastic trajectories [20]. They satisfy the normalization condition $\sum_{n=0}^\infty \sum_{\beta=\pm} P_{\beta,n}(t) = 1$ and obey a master equation resembling Eq. (2):

$$\dot{P}_{\beta,n}(t) = W_{-\beta}(t)P_{-\beta,n-1}(t) - W_\beta(t)P_{\beta,n}(t) \quad (7)$$

for $n \geq 1$ and $\dot{P}_{\beta,0}(t) = -W_\beta(t)P_{\beta,0}(t)$ for $n = 0$. The populations of the states can be obtained as $p_\beta(t) = \sum_{n=0}^\infty P_{\beta,n}(t)$ and the probability $P_n(t)$ to have n -jumps is then $P_n(t) = P_{+,n}(t) + P_{-,n}(t)$. Given $P_n(t)$, any moment $\langle n^k(t) \rangle := \sum_{n=0}^\infty n^k P_n(t)$ ($k = 1, 2, \dots$) can be obtained. Deriving explicit analytical expressions, however, presents a nontrivial task. Fortunately, the first two moments are an exception to this rule. In particular, from Eqs. (7) and (2), it follows that the phase frequency $\Omega_{\text{ph}}(t) := \pi(d/dt)\langle n(t) \rangle$ can be expressed as

$$\Omega_{\text{ph}}(t) = \pi[W_+(t)p_+(t) + W_-(t)p_-(t)]. \quad (8)$$

This result coincides with one obtained earlier in Refs. [18, 19]. For the averaged phase we have $\langle \phi(t, t_0) \rangle =$

$\int_{t_0}^t \Omega_{\text{ph}}(t') dt'$. To obtain $\overline{\Omega}_{\text{ph}}$, one must take the limit $\overline{\Omega}_{\text{ph}} = \lim_{t \rightarrow \infty} \langle \phi(t, t_0) \rangle / (t - t_0)$. For a periodic driving the asymptotic behaviors for both $p_\beta(t)$ and $\Omega_{\text{ph}}(t)$ are periodic functions of time with period \mathcal{T} . Thus, $\overline{\Omega}_{\text{ph}} = (1/\mathcal{T}) \int_0^\mathcal{T} dt \Omega_{\text{ph}}^{(\infty)}(t)$, where $\Omega_{\text{ph}}^{(\infty)}(t)$ is given by Eq. (8) with $p_\beta(t)$ replaced by the asymptotic solution $p_\beta^{(\infty)}(t)$ of the master equation (2), which formally is obtained by letting $t_0 \rightarrow -\infty$.

The calculation of the phase diffusion coefficient $D_{\text{ph}}(t) := (\pi^2/2)(d/dt) [\langle n^2(t) \rangle - \langle n(t) \rangle^2]$ in closed form turns out to be intricate and lengthy, yielding

$$2D_{\text{ph}}(t) = \pi \Omega_{\text{ph}}(t) - 2\pi^2 \delta W(t) \sum_{\beta=\pm} \beta \int_{t_0}^t dt' W_\beta(t') \times p_\beta^2(t') \exp \left[- \int_{t'}^t W(\tau) d\tau \right], \quad (9)$$

where $\delta W(t) = W_+(t) - W_-(t)$ and $W(t) = W_+(t) + W_-(t)$. This remarkable *exact* result in Eq. (9) was originally derived in a different context in Ref. [18] within a slightly different approach. Note that $D_{\text{ph}}(t)$ still depends, like $\Omega_{\text{ph}}(t)$, on the initial time t_0 . This dependence is asymptotically lost in the limit $t_0 \rightarrow -\infty$, where $p_\beta(t)$ is replaced by the asymptotic periodic solution $p_\beta^{(\infty)}(t)$. The evaluation of the mean diffusion coefficient \overline{D}_{ph} proceeds similarly to $\overline{\Omega}_{\text{ph}}$. For the considered case of rectangular driving, both, $\overline{\Omega}_{\text{ph}}$ and \overline{D}_{ph} are evaluated to read:

$$\overline{\Omega}_{\text{ph}} = \frac{\pi W}{2} \left\{ 1 - \delta p_{\text{eq}}^2 \left[1 - \frac{4 \tanh(W\mathcal{T}/4)}{W\mathcal{T}} \right] \right\}, \quad (10)$$

and

$$2\overline{D}_{\text{ph}} = \pi \overline{\Omega}_{\text{ph}} - \frac{2\pi^2}{\mathcal{T}} \delta p_{\text{eq}}^4 \tanh^3(W\mathcal{T}/4) - \frac{\pi^2}{2\mathcal{T}} \delta p_{\text{eq}}^2 (1 - \delta p_{\text{eq}}^2) \left\{ 12 \tanh(W\mathcal{T}/4) - W\mathcal{T} [1 + 2 \text{sech}^2(W\mathcal{T}/4)] \right\}, \quad (11)$$

where W denotes the sum of the forward and backward rates in Eq. (3) for a fixed value of the field \mathcal{E}_0 , i.e., for $\epsilon(t) = \epsilon_0 = e r \mathcal{E}_0$, and $\delta p_{\text{eq}} = \tanh(\epsilon_0/(2k_B T))$ is the absolute value of the difference of the equilibrium populations. The inverse Fano factor of the counting process $R := \pi \overline{\Omega}_{\text{ph}} / (2\overline{D}_{\text{ph}})$ provides a reliable quality measure of synchronization [3, 4].

The quantum features of the so derived synchronization quantities are rooted in the quantum rate expressions entering Eqs. (10) and (11). By analogy with the findings for quantum stochastic resonance (QSR) in symmetric quantum systems [7], one could expect that there is no thermal noise assisted synchronization for $\alpha \leq 1$. Indeed, we could not identify *noise-assisted* synchronization in this parameter regime. In contrast, employing the reasoning put forward for QSR within $\alpha > 1$ in Ref. [8], one then supposes quantum synchronization to emerge in this latter regime.

Indeed, for $\alpha > 1$ the synchronization scenario depends sensitively on the value of the driving frequency. As discussed above, for driving time scales with a frequency much smaller than the tunneling rate at $T = 0$, i.e., $\Omega \ll W_{T=0} = \pi \Delta^2 [2\omega_c \Gamma(2\alpha)]^{-1} [\epsilon_0/(\hbar\omega_c)]^{2\alpha-1} \exp[-\epsilon_0/(\hbar\omega_c)]$, $\overline{\Omega}_{\text{ph}}$ matches the external frequency for sufficiently low temperatures. This is depicted in the upper left panel of Fig. 1, where in addition the behavior of \overline{D}_{ph} is presented. Notice that quantum synchronization at sufficiently low temperature indeed is impressively well accomplished, as it is reflected by the large values of the R -factor (see the lower right panel of Fig. 1). As the temperature increases above a certain critical-like value, however, the R -factor diminishes and the quality of synchronization deteriorates.

With increasing driving frequency such that the condition $\Omega \ll W_{T=0}$ is no longer being met, we enter the regime of thermally induced phase synchronization in the quantum regime, as depicted in the upper right and lower left panels in Fig. 1. We observe a range of temperature where the external frequency and $\overline{\Omega}_{\text{ph}}$ coincide. Moreover, the diffusion coefficient exhibits a pronounced cusp-like minimum. The results plotted in those two panels convincingly illustrate the constructive role of quantum thermal noise for quantum synchronization. Notice that with increasing driving frequency the quality factor associated to the synchronization effect starts to diminish, see the lower right panel in Fig. 1. Moreover, the temperature range where frequency locking is observed shrinks. The quality factor R displays a cusp-like feature similar to the one discussed recently by Park and Lai [21] within the context of noisy, classical synchronization. Upon further increasing the driving frequency, the tunneling rate is too slow compared to the driving frequency for the tunneling dynamics to follow the external oscillations. Thus, quantum synchronization is lost (not depicted).

In conclusion, we have discovered the existence of a quantum stochastic synchronization in an externally driven spin-boson system which undergoes tunneling transitions between two states. We exemplified the phenomenon for non-adiabatic charge transfer in molecular complexes. The estimates of parameter values for an experimental test of our theoretical predictions are: driving frequencies $\Omega \sim 10^{-1} - 10^3 \text{ s}^{-1}$, electrical field strength $\mathcal{E} \sim 10^4 - 10^5 \text{ V/cm}$, and temperatures $T \sim 20 - 100 \text{ K}$, which are readily achieved in the laboratory. The main quantum features of the discussed synchronization phenomenon are robust and they are not critically dependent on the details of the underlying dissipation mechanism. This is so because the phenomenon of quantum synchronization is primarily based on the existence of a low-temperature limit of the tunneling rates. We, consequently, expect our very general results to be useful in other contexts such as for optimizing the noisy dynamics of nano-mechanical systems in the quantum regime [10]. The authors are confident that these riveting findings for quantum synchronization will spur experimental interests for diverse systems that involve a controllable quantum tunneling between distinct quantum states.

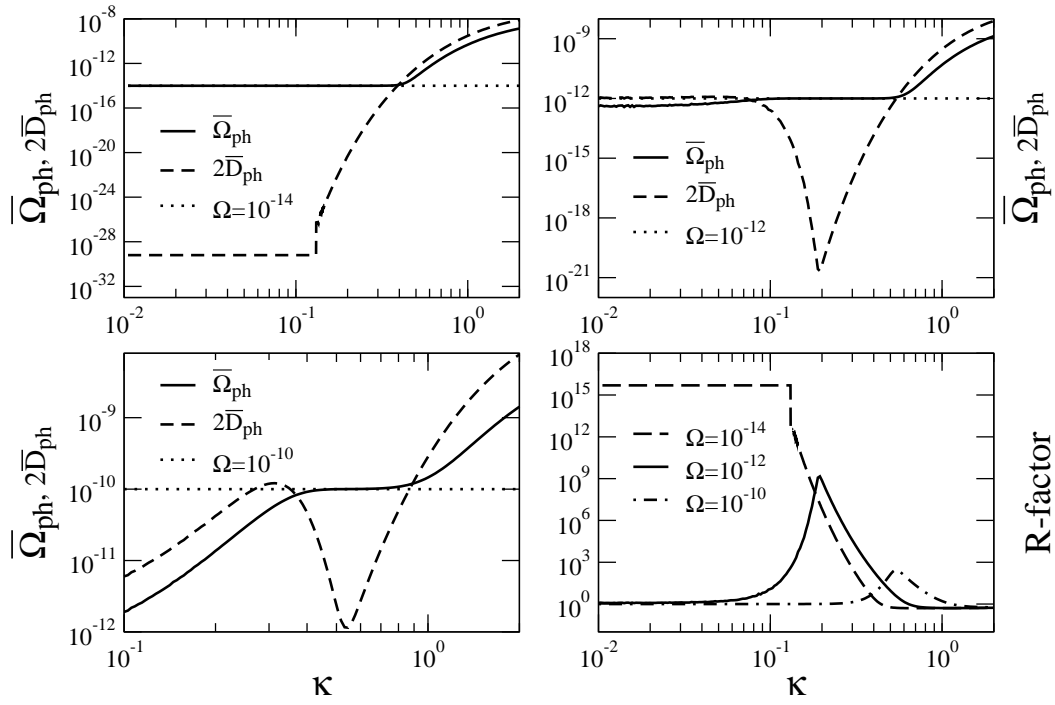


FIG. 1: Mean jump frequency $\bar{\Omega}_{\text{ph}}$, phase diffusion coefficient \bar{D}_{ph} (upper right, upper left and bottom left panels) and the synchronization quality factor R (bottom right panel) versus the scaled temperature $\kappa = k_B T / \hbar \omega_c$ for three values of the driving frequency Ω . The parameter values used are: $\alpha = 10$, $\Delta = 4 \cdot 10^{-4}$, $\epsilon_0 = 5$. Frequencies are scaled with ω_c and energies with $\hbar \omega_c$. For $\hbar \omega_c = 12.5$ meV, $\omega_c \approx 1.9 \cdot 10^{13}$ 1/sec, and $r = 14.9$ Å (taken to match the charge transfer in molecules from Ref. [14]) yields $\mathcal{E}_0 \approx 4.19 \cdot 10^4$ V/cm. The value $\kappa = 1$ corresponds approximately to 145 K.

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